Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.36229

W(VI) Adsorption onto Bacterial Surfaces: Constraints on Adsorption Mechanisms

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Bacterial adsorption of W(VI) can control tungsten bioavailability and mobility in geochemical systems, and represents a potential approach to efficiently remove and recover W from contaminated waters. Our objective was to determine the important adsorption mechanisms in order to enable modeling or optimization of W(VI) adsorption in natural and engineered systems. We measured the adsorption of W(VI) onto a Gram-positive bacterial species, *Bacillus subtilis*, and onto a Gram-negative bacterial species, *Shewanella oneidensis*, as functions of pH, aqueous Zn concentration, and ionic strength, and with and without blocking of either sulfhydryl or amine sites on each type of bacterial surface. We used surface complexation and aqueous speciation modeling to constrain the important W(VI) adsorption behaviors.

W(VI) adsorbs onto each bacterial species, with the extent of adsorption strongly dependent on pH. The concentration of adsorbed W(VI) onto both bacterial species is greatest at low pH, and decreases from pH 4 to 5.5, with negligible adsorption above pH 5.5. At pH 4 in 0.1 M NaCl, *B. subtilis* adsorbs approximately twice the concentration of W(VI) from solution than does *S. oneidensis*. Decreasing ionic strength to 0.001 M NaCl increases the extent of W(VI) adsorption by approximately 20-30% from pH 4.5 – 5.5, and the presence of aqueous Zn decreases the extent of W(VI) adsorption onto *B. subtilis* below pH 4.5, but increases the extent of adsorption above pH 4.5. Blocking experiments indicate that neither sulfhydryl nor amine sites are involved in W(VI) binding onto the bacterial surfaces.

The experimental results, coupled with aqueous speciation modeling and surface complexation modeling, suggest that bacterial adsorption of anionic WO4-2 does not occur to a significant extent unless a divalent cation is present in the system. In the Zn-free systems, W(VI) adsorption is controlled by the binding of neutrally-charged WO30(aq) onto bacterial carboxyl and/or phosphoryl sites. The low pH binding is dominantly electrostatic, likely controlled by interactions between negatively-charged bacterial sites and the polar WO30(aq) molecule. Above approximately pH 4.5, our data suggest that the presence of aqueous Zn+2 leads to ternary complexation involving bridging of WO4-2 and the negatively-charged bacterial surface by Zn+2.